



**On the wintertime
low bias of Northern
Hemisphere CO in
global model studies**

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On the wintertime low bias of Northern Hemisphere carbon monoxide in global model studies

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Received: 17 October 2013 – Accepted: 12 December 2013 – Published: 6 January 2014

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Published by Copernicus Publications on behalf of the European Geosciences Union.

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Abstract

The uncertainties in the global budget of carbon monoxide (CO) are assessed to explain causes for the long-standing issue of Northern Hemispheric wintertime underestimation of CO concentrations in global models. With a series of MOZART sensitivity simulations for the year 2008, the impacts from changing a variety of surface sources and sinks were analyzed. The model results were evaluated with monthly averages of surface station observations from the global CO monitoring network as well as with total columns observed from satellites and with vertical profiles from measurements on passenger aircraft. Our basic simulation using MACCity anthropogenic emissions underestimated Northern Hemispheric near-surface CO concentrations on average by more than 20 ppb from December to April with the largest bias over Europe of up to 75 ppb in January. An increase in global biomass burning or biogenic emissions of CO or volatile organic compounds (VOC) is not able to reduce the annual course of the model bias and yields too high concentrations over the Southern Hemisphere. Raising global annual anthropogenic emissions results in overestimations of surface concentrations in most regions all-year-round. Instead, our results indicate that anthropogenic emissions in the MACCity inventory are too low for the industrialized countries during winter and spring. Thus we found it necessary to adjust emissions seasonally with regionally varying scaling factors. Moreover, exchanging the original resistance-type dry deposition scheme with a parameterization for CO uptake by oxidation from soil bacteria and microbes reduced the boreal winter dry deposition fluxes and could partly correct for the model bias. When combining the modified dry deposition scheme with increased wintertime road traffic emissions over Europe and North America (factors up to 4.5 and 2, respectively) we were able to optimize the match to surface observations and to reduce the model bias significantly with respect to the satellite and aircraft observations. A reason for the apparent underestimation of emissions may be an exaggerated downward trend in the RCP8.5 scenario in these regions between 2000 and 2010, as this scenario was used to extrapolate the MACCity emissions from their base

ACPD

14, 245–301, 2014

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year 2000. This factor is potentially amplified by a lack of knowledge about the seasonality of emissions. A methane lifetime of 9.7 yr for our basic model and 9.8 yr for the optimized simulation agrees well with current estimates of global OH, but we cannot exclude a potential effect from errors in the geographical and seasonal distribution of OH concentrations. Finally, underestimated emissions from anthropogenic VOCs can also account for a small part of the missing CO concentrations.

1 Introduction

Carbon monoxide (CO) is a product of incomplete combustion and is also produced from oxidation of volatile organic compounds (VOC) in the atmosphere. It is of interest as an indirect greenhouse gas and acts as a major sink for the OH radical. The resulting inverse relationship between CO and OH is a reason for its important indirect control function on the global atmospheric chemical composition (Novelli et al., 1998). CO is also a precursor for tropospheric ozone and thus affects regional air quality.

Due to its mean tropospheric lifetime of about two months, CO can be transported globally and is often used as a tracer for long-range pollution transport (e.g. Li et al., 2002; Liu et al., 2003; Duncan and Bey, 2004; Law and Stohl, 2007; Shindell et al., 2008; Drori et al., 2012; Cristofanelli et al., 2013). Tropospheric CO builds up over the Northern Hemisphere (NH) during wintertime, when emissions are high and photochemical activity is low. The surface CO concentrations peak around March with typical mixing ratios measured at background stations of around 150 ppb. Above and downwind of strongly polluted areas in East Asia, North America, or Europe, wintertime concentrations are significantly higher. Values above 1000 ppb have been reported over Eastern China (e.g., Wang et al., 2004; Gao et al., 2005). Summertime concentrations are significantly lower with NH background values going down to 100 ppb due to the faster photochemical depletion of CO. Tropical and Southern Hemispheric concentrations are generally lower (values as low as 35–40 ppb have been observed in remote areas) due to the limited amount of anthropogenic emissions and the large

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ocean surface (Novelli et al., 1998; Duncan et al., 2007). In some regions, the highest concentrations are measured from August to October, largely influenced by emissions from biomass burning (Worden et al., 2013).

The main sources of CO are from anthropogenic and natural direct emissions and from the oxidation of methane and other VOCs. Dominant emissions are from road traffic, fossil fuel and biomass burning together with smaller contributions from vegetation and the oceans. Automobile traffic caused about 85 % of the 2008 emissions of CO from fossil fuels in the USA (<http://www.epa.gov/air/emissions/>) but only 26 % of the 2011 emissions for the European Union (EEA, 2013). The strength of this source has been decreasing since the 1990s in the industrialized countries (e.g. EEA, 2013). There is no clear picture for trends of CO emissions from developing countries, but for East Asia the anthropogenic emissions are estimated to have grown by 34 % from 2000 to 2008 (Kurokawa et al., 2013). Reaction with OH radicals acts as the major CO sink (Prather et al., 2001; Duncan et al., 2007), while deposition in soils contributes about 10–15 % to the global atmospheric CO losses (Conrad, 1996; Sanhueza et al., 1998; King, 1999). A schematic overview of the global CO budget is given in Fig. 1.

In spite of more than 15 yr of research, there are still considerable uncertainties about the global budget of CO: Table 1 lists anthropogenic CO emission totals from recent global and regional emission inventories. The inventories report global anthropogenic emissions of 494–611 TgCO_{yr}⁻¹ for the period 2000–2005. Emission reductions in the industrialized countries since the late 1990s are largely compensated by an increase in the developing countries, particularly in Asia.

The second most important direct CO emission source is biomass burning. Observations of burned area, active fires and fire radiative power from various satellite instruments have been used to constrain these emissions on the global scale. Global and regional totals for the year 2005 from the most recent emission inventories GFEDv2 (van der Werf et al., 2006), GFEDv3 (van der Werf et al., 2010), and GFASv1 (Kaiser et al., 2012) are displayed in Table 2. Note the large differences in continental-scale emissions between the inventories and the large interannual variability in the datasets. Bio-

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genic emission estimates from the widely used MEGAN-v2 emission inventory (Guenther et al., 2006) and the MEGAN-MACC inventory (Guenther et al., 2012) are also listed in Table 2.

5 Global in-situ production from methane oxidation is estimated at about 800 Tgyr^{-1} (Prather et al., 2001). The ensemble model simulations performed for the year 2000 in Shindell et al. (2006) yielded 578 to 999 Tgyr^{-1} of CO from this source with a multi-model mean of 766 Tgyr^{-1} . Since methane had been prescribed as a uniform mixing ratio at the surface for all models, this variability reflects the differences in the OH radical distribution among the models. The chemical production of CO from oxidation of other VOCs is even more uncertain. These emissions are dominated by biogenic
10 sources, notably from isoprene, methanol and terpenes. While Prather et al. (2001) reported 430 Tgyr^{-1} of CO, Duncan et al. (2007) calculated a range of 501 – 542 Tgyr^{-1} for the period 1988–1997 with the GEOS-Chem model. The global simulations performed for Shindell et al. (2006) yielded 547 – 1198 Tgyr^{-1} with a multi-model mean of
15 730 Tgyr^{-1} , pointing to large differences among the VOC emission schemes and again the OH distribution in the various models.

The global annual loss rate of CO by oxidation with OH roughly balances the total sources and is estimated as 1500 – 2700 Tgyr^{-1} (Prather et al., 2001), previous model studies derived 2231 – 2618 Tgyr^{-1} (Duncan et al., 2007 and references therein). This
20 important CO sink depends on an accurate calculation of the global OH distribution, which itself cannot be measured directly. Instead, the global annual average CO loss term can be quantified by the methane lifetime, and evaluated using methylchloroform observations. Prather et al. (2001) estimated a methane lifetime of 9.6 (8.2 – 11.0) yr, while Shindell et al. (2006) calculated a multimodel mean of 9.7 (± 1.7) yr. In a more recent study, Prather et al. (2012) derived a present-day methane lifetime of 9.1 (± 0.9) yr
25 from a systematic exploration of known greenhouse gas budgets and its uncertainties.

The only other known loss process of CO besides reaction with OH is dry deposition. Reported CO dry deposition velocities for vegetated surfaces based on measurements are relatively low compared with other substances and reach from 0 to 0.004 ms^{-1}

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with most values below 0.001 ms^{-1} (King, 1999; Castellanos et al., 2011 and references therein). Prather et al. (2001) reported the global sink to be as high as $250\text{--}640 \text{ Tgyr}^{-1}$. King (1999) confirmed the $190\text{--}580 \text{ Tgyr}^{-1}$ range proposed by Conrad and Seiler (1985) derived from empirical approaches but with a higher probability for the lower values. Based on a constant dry deposition velocity of 0.03 cm s^{-1} Sanhueza et al. (1998) estimated the global gross uptake to be 115 to 230 Tgyr^{-1} . Bergamaschi et al. (2000) derived annual deposition fluxes of about 300 Tgyr^{-1} using the same constant deposition velocity, but setting the deposition velocity to zero in deserts and areas with monthly mean temperatures below 0°C .

Little focus has been placed so far on the role of CO dry deposition in global modelling. Most models apply a dry deposition scheme based on the resistance model of Wesely (1989). Often, the resistance values for O_3 and SO_2 are scaled to derive parameterizations for CO and other substances (Wesely and Hicks, 2000). Such schemes give quite variable dry deposition velocities (Stevenson et al., 2006) but there has not been a systematic intercomparison among the different models yet except for the study of Dentener et al. (2006), where the focus was on wet deposition of NO_x and SO_2 , however. Other models assume that the CO dry deposition losses are to some degree counterbalanced by the plant emissions (e.g. Arellano et al., 2004, 2006; Duncan et al., 2007) and therefore neglect both the soil sink and the biogenic emissions. Only a few models (Emmons et al., 2010; Lamarque et al., 2012) have extended their dry deposition scheme with a parameterization for CO and H_2 uptake by oxidation from soil bacteria and microbes following the work of Sanderson et al. (2003) which itself was based on extensive measurements from Yonemura et al. (2000). In this case, CO and H_2 deposition velocities depend on the soil moisture content of the specific land cover type and on snow cover. Ehhalt and Rohrer (2009) evaluated existing parameterizations of H_2 uptake by soils and found that all existing model studies did not fully account for the essential dependencies of dry deposition velocities on snow cover, soil moisture and vegetation type found in the field experiments except the work from Sanderson et al. (2003).

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As there have been only modestly decreasing trends observed in the concentrations of CO after the 1990s (Fortems-Cheiney et al., 2011; Worden et al., 2013), the sources and sinks of CO must be approximately in balance, except for some seasonal and interannual variations in the CO budget (Duncan and Logan, 2008). The few estimates of the atmospheric CO burden of about 360 Tg (Prather et al., 2001) or 345 (248–407) Tg calculated as the multimodel mean (spread) in Shindell et al. (2006) are similar, even if they apply to different periods.

For a long time, global models have underestimated carbon monoxide concentrations in the NH as is comprehensively shown in the model intercomparison by Shindell et al. (2006). Their multimodel mean from 26 global models exhibited large underestimates of NH extratropical CO, particularly during winter and spring when compared to NOAA GMD station measurements. Such a bias is still existent in more recent model studies like Elguindi et al. (2010), Huijnen et al. (2010), Fisher et al. (2010) and Williams et al. (2013).

With the growing availability of atmospheric CO observation data from surface and in-situ observations as well as from satellites, also a number of global model inversions have been performed to derive optimized CO sources from these observations (Bergamaschi et al., 2000; Pétron et al., 2002, 2004; Arellano et al., 2004, 2006; Müller and Stavrou, 2005; Duncan et al., 2007; Pison et al., 2009; Kopacz et al., 2010; Fortems-Cheiney et al., 2011; Hooghiemstra et al., 2011, 2012; Miyazaki et al., 2012). Similar to the forward-model intercomparison from Shindell et al. (2006) most of these studies found a significant underestimation of CO sources in their a-priori estimates and assigned those to missing emissions in the current inventories. The additional total CO sources needed were as high as 392 Tgyr⁻¹ (Fortems-Cheiney et al., 2011), 492 Tgyr⁻¹ (Kopacz et al., 2010) and 530 Tgyr⁻¹ (Hooghiemstra et al., 2012, including CO sources from VOC emissions). From both forward and inverse model studies, a major part of the missing source was attributed to NH anthropogenic emissions (e.g. Shindell et al., 2006; Müller and Stavrou, 2005; Kopacz et al., 2010; Hooghiemstra et al., 2012).

In the Monitoring Atmospheric Composition and Climate (MACC) project, the technique of variational data assimilation has been employed to perform a global reanalysis simulation for the years 2003–2010 (Inness et al., 2013). Total and partial columns as well as vertically resolved observations of O₃, CO, and NO₂ from multiple satellite sensors have been assimilated. In the case of CO, this information consisted of total column densities from the Measurements of Pollution in the Troposphere (MOPITT) v4 product (2003–2012) and from the Infrared Atmospheric Sounding Interferometer (IASI, April 2008–2012). For the MACC reanalysis the meteorological forecast and data assimilation system IFS (Integrated Forecast System, <http://www.ecmwf.int/research/ifsdocs>) has been coupled to an updated version of the global chemistry transport model MOZART-3 (Model for Ozone And Related Tracers, version 3; Kinnison et al., 2007) in order to build the coupled MACC system MACC-IFS-MOZ (Flemming et al., 2009; Stein et al., 2012). Validation with independent observations as shown in Inness et al. (2013) revealed that, in spite of the extensive satellite data assimilation in the MACC reanalysis tropospheric concentrations of trace gases are still not always satisfactorily modelled. Although data assimilation helped to reduce the CO NH wintertime underestimation that was seen in a control run without data assimilation, CO concentrations near the surface in the MACC reanalysis were still too low. As shown in Fig. 2 the MOZART control simulation without data assimilation exhibited negative biases as large as 35 % on average at around 850 hPa when compared to MOZAIC profiles at airports north of 30° N. This bias could be reduced most effectively for mid latitudes and for model pressure levels above 950 hPa by the assimilation of CO total columns (Fig. 2). Directly at the surface where the influence of the surface fluxes is most important, the assimilation of CO satellite total columns had only a small effect on the modelled concentrations. In contrast, data assimilation appears to lead to a positive bias at altitudes above 300 hPa. Apart from the biases and missing near-surface sensitivity in the satellite retrievals (see also Klonecki et al., 2012) the remaining model bias in the lower troposphere was attributed to missing emissions in the MACCity inventory.

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Motivated by the discrepancy that is often found between simulated and observed CO concentrations, particularly near the surface in the NH during winter, we have conducted a set of sensitivity simulations to shed further light on the uncertainties in the global CO budget. Specifically, this study addresses the following questions:

1. To what extent could direct or precursor CO emissions be underestimated in current global inventories?
2. What is the impact of uncertainties in the dry deposition parameterisation of CO on the global burden and surface concentrations of CO?
3. How far are the modelled CO concentrations controlled by global levels and distribution of the OH radical?

Based on a series of global chemistry transport model simulations we analyze the response of tropospheric CO concentrations to different sets of trace gas surface boundary conditions spanning the uncertainties in our current knowledge about CO emission and deposition.

The outline of the paper is as follows: in Sect. 2 we will describe the model setup for our simulations and present the model sensitivity simulations. Section 3 summarizes the data sets used for evaluation. The results from the sensitivity studies on surface emissions and dry deposition are presented in Sect. 4, followed by our conclusions in Sect. 5.

2 Model description

In the following, the global model setup used for this study will be described. Inspired by the MACC reanalysis model setup, several MOZART standalone simulations have been conducted, which form the backbone of our CO evaluation. The purposes and specifics of these sensitivity simulations are described in Sect. 2.3.

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2.1 MOZART

MOZART as used in the MACC project is based on the MOZART-3 model code (Kinison et al., 2007) which itself is an extension of the tropospheric chemical transport model MOZART2 (Horowitz et al., 2003) to the stratosphere and mesosphere. Some features have been added from MOZART4 (Emmons et al., 2010): most notably a detailed isoprene degradation scheme as described in Pfister et al. (2008) and a description of SO_x and the nitrogen cycle. Other improvements have been introduced with the model version presented in this paper including an update of chemical rate constants as well as improvements in the photolysis lookup-table approach and the parameterization of polar stratospheric clouds. In the following we will present the model set-up used in this study, focussed on the model's lower boundary conditions.

The MOZART global model simulations presented here are driven by meteorological fields from the ECMWF ERA INTERIM reanalysis (Dee et al., 2011) and accomplish a horizontal resolution of 1.875° × 1.875° with 60 hybrid vertical levels from the surface to the mesosphere (resolution T63L60) and a time step of 15 min. The MOZART chemical mechanism consists of 115 species, 71 photolysis reactions, 223 gas phase reactions and 21 heterogeneous reactions. Reaction rates have been updated to JPL-06 (Sander et al., 2006) wherever applicable. Output for tracer concentrations and other model fields are available with a temporal resolution of three hours.

2.2 Surface boundary conditions

A new inventory of global anthropogenic emissions for trace gases and aerosols has been developed in the MACC and CITYZEN projects (Granier et al., 2011), which provides up-to-date estimates for use in global model simulations (MACCcity). These emission estimates are based on the ACCMIP emissions for the year 2000 (Lamarque et al., 2010). The 2000–2010 MACCcity emissions were obtained by using the 2005 and 2010 emissions from the future scenario RCP8.5 (Representative Concentration Pathway): a linear interpolation was applied to obtain the yearly MACCcity emissions.

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RCP8.5 corresponds to a radiative forcing of 8.5 W m^{-2} in the year 2100 given the respective emissions (Moss et al., 2010; van Vuuren et al., 2011; Riahi et al., 2011). A source-specific seasonality developed for the RETRO project (Schultz et al., 2007; <http://retro.enes.org/>) was applied to the emissions. MACCity also includes ship emissions based on Eyring et al. (2010).

Natural emissions are not included in the MACCity inventory. In all MACC simulations biogenic emissions were based on MEGAN-v2 (Guenther et al., 2006) and other natural emissions were taken from the POET project (Granier et al., 2005) and from the Global Emissions Initiative (GEIA). These emissions are meant to be representative for the year 2000.

The MACCity anthropogenic emissions and the additional natural emissions are used as reference emissions for our MOZART 2008 sensitivity simulations which will be presented in the next section and have also been used in the MACC reanalysis and the reanalysis control simulation without data assimilation.

Biomass burning emissions were generated from a preliminary version of the Global Fire Assimilation System (GFAS) developed in the MACC project (GFASv0, Kaiser et al., 2011). This version is very similar to the finally published inventory GFASv1.0 (Kaiser et al., 2012), but with somewhat smaller totals (for CO deviations for global and regional totals are less than 0.5 %). All biomass burning emissions are available with a daily resolution and have been injected into MOZART's lowest model level where they are distributed quickly within the mixing layer by the boundary layer diffusion scheme.

NO_x from lightning in MOZART is dependent on the distribution of convective clouds, following a parameterization of Price et al. (1997). Aircraft emissions of NO_x and CO are also included in the model (Horowitz et al., 2003).

MOZART contains a detailed representation of both wet and dry deposition. Monthly means of dry deposition velocities for 35 species used in MOZART-3 were pre-calculated offline. For MACC the dry deposition fields originate from a monthly climatology derived from a 10 yr nudged simulation with the global chemistry climate model ECHAM5/MOZ (Richter and Schultz, 2011) where an interactive dry deposition scheme

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was implemented according to the resistance model of Ganzeveld and Lelieveld (1995) and Ganzeveld et al. (1998). For some of our sensitivity simulations the CO dry deposition velocity input fields have been exchanged by monthly mean velocity data for the year 2008 following the parameterization of Sanderson et al. (2003) and calculated with the TM5 chemical transport model (Huijnen et al., 2010). This parameterization assumes a strong dependence of the dry deposition on the soil moisture content and the resulting global patterns look strikingly different from those of the original scheme. Figure 3 compares January and July dry deposition fields from both schemes. Deposition velocities according to the Ganzeveld scheme are dependent on air temperature and humidity while the Sanderson scheme generates highest deposition fluxes over the tropics during the rainfall season and for some wet regions in the mid latitudes.

Wet deposition in MOZART is represented as a first-order loss process, with additional in-cloud scavenging and below cloud washout for soluble species (Horowitz et al., 2003). Mixing ratios at the surface are prescribed based on observations for several longer-lived species including methane.

2.3 MOZART sensitivity simulations

For the purpose of analyzing the model sensitivity to CO emission and dry deposition fluxes, we conducted several MOZART simulations for the year 2008 as summarized in Table 3. Our basic simulation MI is based on the surface boundary condition data presented in Sect. 2.2, while the other sensitivity simulations differ in the underlying emission inventories and dry deposition velocities.

The reference simulation MI employs MACCity anthropogenic emissions, together with MEGANv2 biogenic emissions and GFASv0 biomass burning emissions. Basically this configuration was also used for the MACC reanalysis, with some modification for fire emissions (Inness et al., 2013).

Based on simulation MI, we set up seven sensitivity simulations: NH CO concentrations in winter and spring are mostly determined by anthropogenic emissions from traffic and energy consumption. MI+AN tests for the hypothesis that anthropogenic CO

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emissions are underestimated significantly. To incorporate also the anthropogenic VOC precursors, we added simulation MI+VOC with doubled anthropogenic VOC emissions. With respect to natural sources we performed simulation MI+BIO where both biogenic CO and VOC emissions have been doubled. To conclude the sensitivities on the CO emission sources, simulation MI+BB used doubled CO biomass burning emissions from GFASv1. Although these scenarios look quite extreme, they are useful to illustrate the various effects of the emission sectors on the global and regional CO concentrations and their annual variations. Furthermore, uncertainty levels of emissions, particularly for less developed countries and for natural emissions, are still high (e.g. Lamarque et al., 2010).

Simulation MI-DEP tests for the alternative dry deposition velocity scheme from Sanderson et al. (2003). Finally, our optimized simulation MI-OPT applied a modified CO traffic emission scaling for North America and Europe only which exhibits a seasonal variation as shown in Table 4. This simulation utilized the same dry deposition parameterization as in MI-DEP.

All model simulations were analyzed for the year 2008 but started at 1 July 2007 to allow for a spinup period. The model was run at T63LR resolution ($1.875^\circ \times 1.895^\circ$) and with 60 vertical levels from the surface up to 0.1 hPa. Tracer initial conditions were taken from the MACC reanalysis which included assimilation of CO, O₃, and NO₂ satellite information. It should be noted that 2008 was within the three years with lowest atmospheric CO concentrations out of the 13 yr 2000–2012 where satellite observations were available (Worden et al., 2013).

3 Data sets used for evaluation

3.1 Surface station data

Long term measurements of key atmospheric species from surface observation sites around the globe are collected at the World Data Centre for Greenhouse Gases (WD-

CGG) operated by the WMO GAW programme (<http://ds.data.jma.go.jp/gmd/wdcgg/>). In total we used monthly means of CO concentrations measured from 67 surface stations which provided data for all months in 2008, their positions on the globe are shown in Fig. 4 (red diamonds). The observation sites are operated by NOAA/ESRL (USA), CSIRO (Australia), Environment Canada, JMA (Japan), CHMI (Czech Republic), UBA (Germany), EMPA (Switzerland), RIVM (Netherlands), and SAWS (South Africa). We discarded mountain stations and stations which did not deliver monthly means for all months in 2008. Data was averaged over continental-scale areas to minimize the influence from erratic or locally determined measurements. For evaluation we built area-averaged monthly means from all NH and SH stations as well as from those inside the regions defined in Fig. 4 (blue boxes).

3.2 MOZAIC profiles

The MOZAIC (Measurements of Ozone and water vapor by Airbus In-service airCRAFT) program collects ozone, CO, odd nitrogen (NO_y) and water vapor data, using automatic equipment installed on-board several European long-range passenger airliners flying regularly all over the world (Marengo et al., 1998). Here we use CO data from vertical tropospheric transects taken in the vicinity of airports during ascent and descent. For more details of the data processing see Elguindi et al. (2010). In 2008, only three aircraft were operating with the MOZAIC instrumentation and therefore only a limited number of airports were visited regularly (13 airports with a total of 935 profiles). For the evaluation of the 2008 model simulations, we chose tropospheric data from the European airports of Frankfurt (Germany), London (UK), where 300 (80) profiles were available as well as an area average composed of the airports of Atlanta, Dallas, Philadelphia, Portland (OR) in the United States and Toronto and Vancouver in Canada with 219 profiles altogether. This area average is meant to be representative for the North American Area limited by the blue box in Fig. 4. There were no data over East Asia in 2008. We marked all airports used for evaluation with turquoise triangles

in Fig. 4. The model results were interpolated to the times and locations of the MOZAIC observations, afterwards the single profiles were combined to monthly means.

3.3 MOPITT atmospheric columns

The MOPITT (Measurement Of Pollution In The Troposphere) instrument is flying on board the NASA Terra satellite and has been operational since March 2000 (Drummond and Mand, 1996; Deeter et al., 2004). It measures upwelling radiation in the thermal infrared spectral range using gas-filter correlation radiometry. At nadir view, MOPITT offers a horizontal resolution of $22\text{ km} \times 22\text{ km}$ and allows a global coverage in two to three days. MOPITT data used in this study correspond to the daytime CO total columns from the level 2 version 5 product. MOPITT version 5 offers CO data retrieved in both near-infrared (NIR) and thermal-infrared (TIR) radiances (Deeter et al., 2013). The MOPITT products are available through the following web server: <http://www2.acd.ucar.edu/mopitt/products>. For evaluation we use the multispectral CO product which is based on both NIR and TIR radiances. Exploiting TIR and NIR radiances together increases the sensitivity to CO in the lowermost troposphere (Worden et al., 2010; Deeter et al., 2011, 2012). The observations are area-averaged monthly means from the regions defined in Fig. 4 (blue boxes). The modelled CO total columns were calculated by applying the MOPITT averaging kernels. Details about the method of calculation are given in Deeter et al. (2004) and Rodgers (2000).

3.4 IASI atmospheric columns

IASI (Infrared Atmospheric Sounding Interferometer) is a nadir looking remote sensor, launched in 2006 on the polar orbiting MetOp-A satellite. IASI has a twice daily global coverage, with 4 pixels of 12 km diameter (at nadir) and a 2200 km swath width. Its spectral resolution is 0.5 cm^{-1} (apodized) with continuous spectral coverage from 645 to 2760 cm^{-1} that allows column and profile retrievals of several different trace gases (Clerbaux et al., 2009). For carbon monoxide, profiles are retrieved using an optimal

estimation approach, implemented in the Fast Optimal Retrievals on Layers for IASI (FORLI) software (Hurtmans et al., 2012). The spectra are processed in near real time and the CO profiles are distributed with error covariance, averaging kernels and quality flags information. The IASI CO product was validated against ground based observations (Kerzenmacher et al., 2012), aircraft data (Pommier et al., 2010; Klonecki et al., 2012) and satellite measurements (George et al., 2009). The FORLI-CO products are available through the Ether French atmospheric database website: www.pole-ether.fr. The IASI column values used here are daytime data that were processed with version 20100815 of the FORLI-CO retrieval algorithm. For this study area-averaged monthly means from the regions defined in Fig. 4 (blue boxes) are used.

4 Results

4.1 MOZART sensitivity simulations

In the following, we use MOZART sensitivity simulations to identify the potential role of the various CO surface fluxes for the underestimation of the near-surface concentrations. First we compare our basic simulation (MI), using MACCity anthropogenic and natural emissions, to observations gathered from the World Data Center for Greenhouse gases (WDCGG) database. Figure 5 shows monthly means of surface station CO mixing ratios and of near-surface model concentrations interpolated to the station location for 67 stations ordered by latitude. Note that all mountain stations as well as those stations which did not deliver monthly means for all months in 2008 have been excluded.

The station data exhibits the highest values and also the largest variability at northern mid latitudes (30–60° N) during wintertime with mean January concentrations of more than 300 ppb for three stations, namely Hegyhatsal (Hungary), Payerne (Switzerland) and Black Sea (Romania). In addition, Payerne exceeded this threshold in February 2008 and the station Tae-ahn (South Korea) reached monthly mean concentrations

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of more than 300 ppb for March to May 2008 (not shown). NH concentrations are generally higher in winter (and spring) than in summer. The station observations in the Southern Hemisphere (SH) show a relatively uniform distribution both in winter and summer with background concentrations of about 40–70 ppb. Modelled NH CO mixing ratios are mostly lower than the observations, particularly during wintertime. In January, the model underestimates the concentrations for the arctic stations by about 50 ppb. In the SH model and observations match better, with an overestimation of up to 10 ppb being typically seen. The global distributions of atmospheric CO can be derived with high spatial resolution from the satellite total column observations. As shown in Fig. 6, the MOPITT and IASI products agree well for most regions. MOPITT total columns are somewhat higher than IASI for several NH regions in January, but IASI attains a better spatial coverage. The MI simulation is able to reproduce the general features and levels of the satellite CO distribution, but calculates higher CO columns over India and slightly lower columns for other polluted regions like Europe, North America and East Asia. The North African biomass burning regions in January are less pronounced than for the satellites, while there seems to be an overestimation from the model over SH tropical West Africa in July.

Following the findings from the MACC reanalysis and from the previous global studies, the negative model bias in boreal winter and spring CO concentrations could be explained by either missing sources from surface emissions or by an overestimation of the surface dry deposition sink. Thus we expanded our basic model simulation for the year 2008 with a suite of sensitivity simulations covering the potential sources of uncertainty in the surface fluxes (emissions and deposition) as presented in Sect. 2.4. Impacts from uncertainties in the modelling of OH on the CO budget will be discussed subsequently. Modelled tropospheric CO mixing ratios and total column densities are evaluated with the observational data introduced in Sect. 3.

4.1.1 Emissions

First, we compare monthly means of near-surface model concentrations with monthly mean surface station data from the WDCGG database averaged over the Northern and Southern Hemisphere as well as for the regions Europe, North America and East Asia.

Model data was interpolated to the station location. In Fig. 7 the model bias is shown for the base simulation MI (MACCity emissions) and for the sensitivity simulations MI+AN, MI+BIO, and MI+BB.

The simulation with MACCity emissions (MI) underestimates the observed surface concentrations in the NH in all seasons except summer, with low biases exceeding 20 ppb from December to April and exceeding 40 ppb in January. The largest underestimation of about 75 ppb is found over Europe. During boreal summer, the modelled CO concentrations match on average the observations, with a slight underestimation for Europe and overestimation for North America. NH summertime concentrations are more influenced by biogenic and biomass burning emissions, and the photochemical sink via reaction with the OH radical plays a larger role.

East Asian observed concentrations agree relatively well with the MI simulation for the whole year, at least for the four surface stations which could be taken into account here. This relative compliance is probably due to the recently updated regional emission information and to the better emission predictions for China in the MACCity inventory (Riahi et al., 2011). For the sake of completeness we also report here the good agreement between simulated and observed SH concentrations, only a small positive model bias of 5 to 10 ppb is found all year round.

Similarly to the surface concentrations, the modelled CO total columns are compared with the observed total columns from MOPITTv5 and IASI (Fig. 8). The satellite observations are more representative for the free troposphere and accomplish a substantial coverage over the regions of interest. Due to the differences in spatial sampling between the two instruments, and the different methods used to obtain the CO total column product, a significant spread between the two satellite products of up to

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9 % averaged over the NH exists (see also Worden et al., 2013). Over the NH, IASI observed lower total columns than MOPITT from October to March and higher total columns in July/August. The findings from the surface station evaluation of the MI simulation generally hold true also for the total columns with a few differences: MI always underestimated NH MOPITT total columns as well as the IASI columns from January to October. Largest biases occur for MOPITT – MI in March with 18 % (NH), 24 % (Europe) and 22 % (North America).

As expected, in the extreme sensitivity simulation with doubled anthropogenic emissions (MI+AN, see Figs. 7 and 8), the CO concentrations overestimated the observed values in all regions. However, deviations from the observations are smaller than in the MI simulation from January to March over the NH which can mostly be attributed to the North American and European regions. The overestimation is less pronounced during the summer months, when the CO loss due to reaction with OH gains more importance.

The simulation MI+BIO (doubled biogenic CO and VOC emissions) enhanced the modelled surface CO concentrations by 20 to 40 ppb for all regions. Since the biogenic emissions mainly occur over the tropics and the NH landmasses with a summer maximum, we find that in summer, the emissions are overestimated over the whole NH. At higher altitudes, this summer overestimation seems to last until winter 2008/2009 (Fig. 8). SH concentrations are biased high during all months for both surface concentrations and total columns. The MI+BB simulation (doubled CO biomass burning emissions) leads to overestimation of the peak surface concentrations during NH spring and summer, as can be noticed in the wildfire-influenced regions of North America and East Asia. When compared with IASI observations, the MI+BB simulation performs better than the other runs. This is as expected, since IASI is more sensitive to transient biomass burning events, which are also included in our simulation (Worden et al., 2013). Both simulations with enhanced biogenic or wildfire emissions are not able to improve the negative bias seen by the models in NH winter without introducing significant overestimations during boreal summer and over the SH. This constrains the

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contribution of biases in biogenic or biomass burning emissions to the modelled NH wintertime low bias in CO.

To account for the uncertainties in the emissions of anthropogenic CO precursors, we also ran MOZART using MACCity emissions together with doubled VOC anthropogenic emissions (MI+VOC). These increased emissions are able to enhance wintertime and spring surface CO concentrations by 5 to 15 ppb (Fig. 9, grey lines) and could therefore add a small portion to the missing NH CO concentrations with the greatest improvement seen in spring.

4.1.2 Dry deposition

Surface layer mixing ratios from the sensitivity simulation MI-DEP, which used the parameterization by Sanderson et al. (2003) for deposition velocities, together with the MACCity emissions, are displayed in Fig. 9. When compared with MI, the reduced deposition fluxes over the NH landmasses result in higher concentrations of around 10 ppb in summer and 20 ppb in winter when OH concentrations are low. Over Europe, where the differences between the two schemes are quite large, wintertime concentrations are higher by 30 ppb whereas values for May to September match the observations very well. Concentrations over East Asia are moderately increased (less than 20 ppb all-year-round), while over North America the summer and autumn values are biased high compared with the observations. Over the SH, the simulation MI-DEP is able to decrease the tropospheric concentrations of CO by up to 5 ppb (August–October) which may be due to the increased deposition fluxes over Africa and South America (see Fig. 9). Overall the use of the parameterization by Sanderson et al. (2003) improved the model results significantly but alone is not able fill the gap to boreal wintertime CO observations, particularly over Europe.

4.1.3 Optimized approach

As indicated in Sect. 2, the MACCity inventory is based on the ACCMIP emissions for the year 2000. Lamarque et al. (2010) pointed out that for USA, Europe, and East Asia published regional inventories had been given preference, notably EPA data for USA, EMEP data for Europe and REAS data for East Asia. Based on a comparison between a fuel-based and a mileage-based emission inventory for one US city, Parrish (2006) concluded that the reason for a high bias in the CO to NO_x ratios in the US National Emission Inventory (NEI) was most likely due to an overestimate of the CO emissions rather than an underestimate in the NO_x emissions and suggested an overestimate of CO emissions (by a factor of 2) in the EPA-2004 data, which also holds true for the more recent EPA estimates up to 2008 (Miller et al., 2008; Lamarque et al., 2010; Granier et al., 2011). EPA also reported on air pollution trends of US cities between 1990 and 2008 and found maximal decadal decreases in CO pollution for US cities of 60 to 80 % for 1990 to 2008 (Riahi et al., 2011). Consequently, in the RCP8.5 scenario an exposure-driven spatial algorithm for the downscaling of the regional emission projection has been employed leading to the highest emission reduction of up to 80 % per decade in those grid cells with the highest exposure for regions where emissions are reduced due to air pollution measures, e.g. USA and Europe. For regions with increasing emissions, (e.g. in Asia) emissions increase proportionally to the acceleration of the economic activity (Riahi et al., 2011). Road traffic provides a major part of the anthropogenic CO emissions in the industrialized countries. Pouliot et al. (2012) compared regional emission inventories for Europe and North America for 2005 and reported 85 % of North American anthropogenic CO emissions and 43 % of European emissions to origin from mobile sources. In Fig. 10 the development of CO anthropogenic emissions 2000–2050 in the RCP8.5 scenario is shown. Although RCP8.5, which is used in MACCity, is a scenario with relatively high greenhouse gas emissions and global warming potential, the CO anthropogenic emissions decrease slowly in the first decade 2000–2010, mostly driven by strong emission reductions in the OECD coun-

tries. Traffic emissions, which build the majority of anthropogenic CO emissions from these countries decrease by 46 % (from 119 Tgyr⁻¹ in 2000 to 44 Tgyr⁻¹ in 2010). For Asia, total emissions are estimated to increase by 10 % until 2020 with largest emission growth rate from industrial emissions and other emission sectors stagnating.

Our optimized simulation MI-OPT tests for the hypothesis that CO emissions from cars are not considered adequately in the MACCity estimate: estimates of automobile emissions are based on defined driving cycles which are lacking of typical short trips, when much more CO is emitted under cold engine conditions (Parrish, 2006; Kopacz et al., 2010; Klemp et al., 2012). This would effect a significant underestimation of traffic CO emissions in the emission inventory, which is potentially amplified by an unrealistic emission reduction 2000–2010 in MACCity. We note, that our simulations don't allow the attribution of the missing CO source to the traffic sector, although (as indicated above) there are reasons to believe that this sector at least contributes to the problem. In reality it may well be a combination of underestimates from various sectors, many of which are poorly constrained. One such sector, which is particularly relevant during winter, is household wood burning (H. Denier van der Gon, personal communication, 2012). Clearly, a more systematic assessment of emission uncertainties would be helpful.

For the simulation MI-OPT we applied a refined scaling to the CO road traffic emissions with monthly scaling factors for Europe and North America only as described in Table 4. Global total added emissions from this simulation are as low as 19 Tgyr⁻¹, compared to 586 Tgyr⁻¹ from simulation MI+AN (Table 3). Applying the modified scaling factors enhances wintertime CO emissions only for these regions and also accounts for realistic CO concentrations from long-range transport. Simultaneously, MI-OPT takes into account the improved dry deposition parameterization by Sanderson et al. (2003) as in MI-DEP.

The simulation MI-OPT is compared with surface observation as shown in Fig. 9. The simulation is able to effectively shift wintertime CO concentrations to higher values for Europe and North America without significantly increasing the CO concentrations

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in East Asia. Applying the scaling factors given in Table 4 results in an almost perfect match to the GAW station observations for Europe and on the NH average. The same holds true over North America for the months December to May. Where concentrations with the MI emissions were already high biased, as for North America in summer or for East Asia in spring and autumn, these overestimations can be attributed to natural emissions. This can be deduced from the MI+BB and MI+BIO simulations, where concentrations for these regions are enhanced similarly.

The comparison of total CO columns from MI-OPT to MOPITT and IASI satellite observations is shown in Fig. 11. NH mean columns are enhanced by 6–9% with respect to the MI simulation, but not enough to catch up with the satellite observations, except for October to December 2008. Similar changes can be seen for the regional averages, with largest increase over Europe (8–12%) and somewhat lower differences for North America (7–10%) and East Asia (7–9%). In summer and autumn, most of the enhancement is due to the changes in the dry deposition parameterization, while in winter and spring the increased emissions dominate the optimization (not shown). CO concentrations over the SH remain mostly unchanged with respect to the simulation MI.

The evaluation results from surface stations and total column observations are complemented by the comparison with MOZAIC profiles over the airports of Frankfurt (Germany) and London (UK) and a composite of 6 airports over North America (Fig. 12). In January 2008 simulation MI is biased low with maximal differences in the lowest model level (ca. 100 ppb over Frankfurt, 55 ppb over London and 80 ppb for North American airports). The model underestimates CO throughout the tropopause with smaller biases as the upper troposphere is approached. In July, MI shows a negative bias of 20–40 ppb throughout the lower troposphere. CO concentrations from the simulation MI-OPT are generally higher than those from MI, thus reducing the bias with respect to the observations. In January the negative bias is reduced to 5–30 ppb near the surface, but may still reach up to 40 ppb in some parts of the free troposphere, whereas in July, MI-OPT concentrations are consistently 10–15 ppb higher than those from MI.

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Our optimization approach illustrates that regionally missing anthropogenic emissions together with modifications in the dry deposition parameterization could explain the observed biases with respect to American and European GAW station observations. Total CO column densities and tropospheric profiles of CO concentrations were also improved. The optimization approach goes some way to addressing the under-estimation of the CO concentrations evident at the MOZAIC airports. The remaining biases point to some model deficiencies in vertical exchange or the OH distribution and may also be due to the coarse resolution of the model being unable to represent strong local sources of pollution at the airport locations. The strong seasonality in the emission scaling factors used for MI-OPT and its regional differences (Table 4) indicate that a more detailed approach is needed to account for the seasonality in global emission inventories, particularly for emissions of anthropogenic origin.

How the modification of wintertime NH CO emissions affects other trace gases is shown for simulation MI-OPT in Fig. 13. First we show the differences in the CO distributions compared with the simulation MI-DEP (top panel): European and North American surface concentrations are enhanced by up to 170 ppb in December. Long-range transport of the additional emissions is confined to the NH with little effect on Asia and the Pacific. In July, increases are always lower than 5 ppb and therefore not shown.

The regionally scaled emissions have only a small impact on the simulated near-surface ozone concentrations (Fig. 13, middle panel) and on the free-tropospheric OH fields (lower panel). Surface ozone increases almost globally, but nowhere by more than 0.5 ppb. This change is smaller than the model bias usually found when MACC results are evaluated with ozone observations (Inness et al., 2013). The OH radical mixing ratios are evaluated at 700 hPa to represent an average tropospheric level and distribution. As expected, OH is reduced in the simulation with scaled MACCity emissions due to the increased sink term by CO oxidation. Differences are up to 3 % in the mid latitudes during winter, where OH concentrations are very small (see average latitudinal profiles on the bottom panel of Fig. 13). OH concentrations in the tropics and

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in the summer hemisphere are generally affected by less than 1 %, suggesting that the CO+OH loss term is marginally changed by modifying the emissions.

We assess the mean OH concentration in the MOZART model, as this would influence the CO budget both for the source term via the chemical conversion of hydrocarbons and for the chemical loss term. OH in the model cannot be evaluated directly, as atmospheric measurements are lacking. A widely used measure for the integrated tropospheric OH concentrations is the methane lifetime $\tau[\text{CH}_4]$. In this study the methane lifetime is 9.7 yr for the MI simulation and 9.8 yr for our simulation MI-OPT. These values are both very close to $\tau[\text{CH}_4] = 9.1(\pm 0.9)$ yr derived by Prather et al. (2012) and to the mean $\tau[\text{CH}_4]$ calculated from the models in Shindell et al. (2006) ($\tau[\text{CH}_4] = 9.72$ yr (6.91–12.38)), indicating that our mean MOZART OH concentration is adequately simulated and the OH sink is not greatly overestimated. As shown here, also the influence of the modified surface fluxes on OH is small. From the evaluation with MOPITT and IASI total columns and MOZAIC profiles however, a prevalent underestimation of free tropospheric model CO concentrations could be deduced. If and how such an underestimation is connected to unrealistic OH distributions or vertical exchange processes in the model, cannot be addressed further without additional observational constraints. Modifications to the chemistry mechanism, which are able to affect global OH and its spatial distribution, such as proposed by Mao et al. (2013), could also result in higher CO concentrations. However, this is not yet well established, as there are large uncertainties related to the magnitude of proposed reaction pathways. Therefore these changes are not considered here.

Finally, the global CO burden calculated from our basic simulation MI is 351 Tg, which is well in the range of reported values in the literature. This was increased to 369 Tg for MI-OPT. 14 Tg out of this 18 Tg increase can be appointed to the reduced dry deposition fluxes in the Sanderson scheme. Also the interannual variability of the CO burden has been calculated with a similar range (Duncan and Logan, 2008).

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Despite the developments in the global modelling of chemistry and of the parameterization of the physical processes, CO concentrations continue to be underestimated in NH winter by most state-of-the-art model simulations. Even with extensive data assimilation of CO satellite retrievals, as it is applied in the MACC forecasts and reanalysis, a significant low CO bias in the lower troposphere remains. Such a bias can in principle originate from either an underestimation of CO sources or an overestimation of its sinks. We addressed both the role of surface sources and sinks with a series of MOZART model sensitivity studies for the year 2008 and compared our results to observational data from ground-based stations, satellite observations, and tropospheric profiles from MOZAIC.

Our basic simulation employed the global emission inventories MACCity, GFASv1 and MEGANv2, of which MACCity and GFASv1 have been developed in the MACC project. Surface CO concentrations as measured by GAW stations averaged over the NH were underestimated by more than 20 ppb from December to April with a maximal bias of 40 ppb in January from this simulation. The bias was strongest for the European region (75 ppb in January) but also apparent over North America, suggesting that wintertime emissions for these regions are missing in the inventories. Modelled concentrations over East Asia were in better agreement with the surface observations, probably reflecting the efforts which were made to update the CO emissions to the recent economic developments, namely in China. Negative biases were also found for total CO columns when evaluated against MOPITT and IASI observational data. Our comparison showed significantly lower NH modelled columns for almost the whole year. MOPITT and IASI products themselves differed by up to 9 % averaged over the NH so a quantification of the total column model bias remains uncertain. As pointed out by Worden et al. (2013) the spread in CO column values across the satellite instruments is mainly due to spatial sampling differences. The modelled CO concentrations were additionally evaluated with MOZAIC profiles over several airports in Europe and

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North America. Tropospheric CO concentrations were mostly underestimated from the model with largest biases for NH winter near the surface.

Four rough sensitivity simulations, defined by doubling one of the original CO sources helped us to identify major sectors which could contribute to the CO underestimation.

5 Doubling the global anthropogenic CO emissions increased the concentrations all year round but predominately during NH wintertime. Nevertheless, concentrations simulated with this extreme scenario were always too high when compared to the surface observations and also for regions where the bias of the original simulation was already small. A sensitivity study accounting for missing anthropogenic VOC emissions showed that
10 although these indirect emissions contribute only about 8 % to the CO sources, their relative impact is higher in winter when biogenic emissions are low. Doubling these emissions increased NH wintertime near-surface concentrations by 5 to 15 ppb on the global and regional scale, hence biases in anthropogenic VOC emissions can account for a part of the missing model CO. A simulation with doubled biogenic emissions, both
15 from CO and the VOCs, led to unrealistically high concentrations during boreal summer and early autumn while SH tropospheric CO was greatly overestimated. Similarly, doubling the direct CO emissions from biomass burning led to CO overestimation peaks during the major NH wildfire seasons in late spring and summer, hence also excluding biomass burning as the major contributor to the wintertime NH CO bias.

20 The influence of the dry deposition sink on the global CO budget has been neglected so far by most of the studies on model inter-comparison and source inversion. Although this sink is small compared with the photochemical sink on global, annual average, it becomes more important over the continents and during the winter months when OH concentrations are low. A sensitivity simulation with a modified CO
25 deposition scheme following Sanderson et al. (2003) reduced the global CO sink by 68 Tgyr^{-1} with strongest reductions over Europe in wintertime. The resulting NH surface CO concentrations were enhanced by 20–30 ppb in winter and by 10–15 ppb in summer with largest increase over wintertime Europe. Nevertheless, the reduced dry deposition alone was not able to explain the total CO bias.

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A simulation which optimally reflects the surface observations was constructed through application of the modified dry deposition together with a seasonally depending scaling of CO traffic emissions for Europe and North America only. With this simulation we tested for a possible underestimation of CO emissions from the traffic sector in the industrialized countries, which could originate from vehicle engines under cold start conditions. While the resulting surface concentrations were very close to the global station observations, also a considerable improvement was reached in comparison to satellite observations and MOZAIC profiles.

In conclusion, our simulations have shown that the mismatch between observed and modelled concentrations of CO during NH wintertime, can be explained by a combination of errors in the dry deposition parameterisation and the following limitations in the emission inventories: (i) missing anthropogenic CO emissions from traffic or other combustion processes, (ii) missing anthropogenic VOC emissions, (iii) a poorly established seasonality in the emissions, (iv) a too optimistic emission reduction in the RCP8.5 scenario underlying the MACCity inventory. There is no indication that our results were greatly influenced by unrealistic global OH levels, but modelled CO concentrations depend on the modelled OH distributions, which are themselves subject to limitations in the chemical mechanisms and the lack of knowledge on seasonality and global patterns of OH.

Taking into account that other recently developed global and regional anthropogenic emission inventories (EDGARv4.2, EMEP, EPA) estimate similar low CO emissions for Europe and North America it remains necessary to question the inventory building process, particularly for the important and highly uncertain traffic sector. Furthermore, research efforts are also needed to improve the estimates for the seasonal variability of anthropogenic emissions. Finally, in future model studies on the inversion of atmospheric CO observations the dry deposition parameterization and its global sink term should be more explicitly documented.

Acknowledgements. This study was funded by the European Commission under the framework programme 7 (contract number 218793). Data from the emission inventories used

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in this study are freely available on the GEIA emission data portal <http://eccad.sedoo.fr/>. IASI has been developed and built under the responsibility of the Centre National d'Etudes Spatiales (CNES, France). We are grateful to Pierre Coheur and Daniel Hurtmans (ULB) for developing the FORLI-CO IASI processing code and to Juliette Hadji-Lazaro (LATMOS) for establishing the IASI MACC near real time processing chain. The authors acknowledge the strong support of the European Commission, Airbus, and the airlines (Lufthansa, Air-France, Austrian, Air Namibia, Cathay Pacific and China Airlines), who carry the MOZAIC or IAGOS equipment and have performed the maintenance since 1994. MOZAIC is presently funded by INSU-CNRS (France), Météo-France, CNES, Université Paul Sabatier (Toulouse, France) and Research Center Jülich (FZJ, Jülich, Germany). IAGOS has been, and is additionally, funded by the EU projects IAGOS-DS and IAGOS-ERI. The MOZAIC-IAGOS data are available via CNES/CNRS-INSU Ether web site <http://www.pole-ether.fr>. ECMWF and Forschungszentrum Jülich are acknowledged for providing supercomputer time, and we thank Claire Granier and Hugo Denier van der Gon for helpful discussions.

The service charges for this open access publication have been covered by a Research Centre of the Helmholtz Association.

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Table 1. Global and regional CO anthropogenic emission totals from recent bottom-up inventories in Tgyr⁻¹. All data except from EPA and Environmental Canada are available online from the Emissions of atmospheric Compounds & Compilation of Ancillary Data (ECCAD) database (<http://eccad.sedoo.fr/>).

	ACCMIP ^a	RETRO/REAS ^b	MACCity ^c	EDGAR v4.2 ^d	EMEP ^e	TNO-MACC ^f	EPA + Env. Canada ^g	REAS v1 ^h
Release year	2010	2008	2010	2011	2007	2009	2013	2007
Reference year	2000	2000/2003	2005/2008	2005	2005	2005	2005	2003/2005
Global	611	577	583/586	494				
Europe	59	49	49/43	46	51	50		
North America	118	70	70/62	82			82	
Asia	320	362	341/352	256				338/345
China	121	158	138/146	90				158/162
Africa	83	69	95/101	61				
South America	22	24	24/24	46				
Oceania	6	3	3/3	4				

^a Lamarque et al. (2010)

^b Schultz et al. (2007)

^c Granier et al. (2011)

^d Janssens-Maenhout et al. (2010)

^e Amann et al. (2005)

^f Denier Van der Gon et al. (2011)

^g <http://www.epa.gov/ttn/chief/eiinformation.html>; <http://www.ec.gc.ca/indicateurs-indicators/>

^h Ohara et al. (2007)

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Table 2. Global and regional 2005 CO emission totals from recent biomass burning and biogenic emission inventories in Tgyr^{-1} . Data are available from the ECCAD database.

	Biomass burning			Biogenic	
	GFEDv2	GFEDv3	GFASv1	MEGANv2	MEGAN-MACC
Release year	2006	2010	2012	2009	2012
Reference year	2005	2008	2008	2000	2008
Global	416	277	325	76	84
global range	369–599 ^a	252–595 ^b	305–404 ^c		83–93 ^d
Europe	19	38	66	5	4
North America	19	16	20	9	7
Asia	82	22	41	18	15
Africa	183	152	140	18	27
South America	101	37	48	19	22
Oceania	11	11	12	7	8

^a 1997–2005

^b 1997–2010

^c 2003–2011

^d 2000–2010

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Table 3. Description of MOZART 2008 sensitivity simulations.

simulation	Anthropogenic emissions	Biogenic emissions	Biomass burning emissions	Dry deposition	Global total CO source added to MI in Tgyr ⁻¹
MI	MACCity	MEGANv2.0	GFASv1	ECHAM5/MOZ	–
MI+AN	MACCity, anthropogenic CO emissions doubled	MEGANv2.0	GFASv1	ECHAM5/MOZ	586
MI+VOC	MACCity, anthropogenic VOC emissions doubled	MEGANv2.0	GFASv1	ECHAM5/MOZ	84
MI+BIO	MACCity	MEGANv2.0, biogenic CO and VOC emissions doubled	GFASv1	ECHAM5/MOZ	535
MI+BB	MACCity	MEGANv2.0	GFASv1, CO biomass burning emissions doubled	ECHAM5/MOZ	323
MI-DEP	MACCity	MEGANv2.0	GFASv1	Sanderson et al. (2003)	68
MI-OPT	MACCity, refined traffic CO scaling for North America and Europe only	MEGANv2.0	GFASv1	Sanderson et al. (2003)	87

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Table 4. Monthly scaling factors for CO traffic emissions over North America and Europe as applied in simulation MI-OPT.

	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
North America	1.25	1.5	2	1.33	1	1	1	1	1	1	1	1
Europe	4.5	3.33	3.33	3	1.5	1	1	1.25	1.5	2	2.5	2.5

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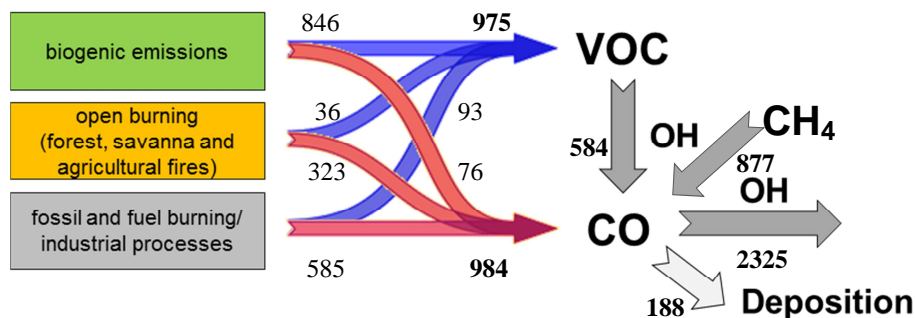


Fig. 1. The global CO budget: sources and sinks. Numbers are in Tgyr^{-1} as estimated from this study using MACCity/MEGAN/GFAS emissions for the year 2008.

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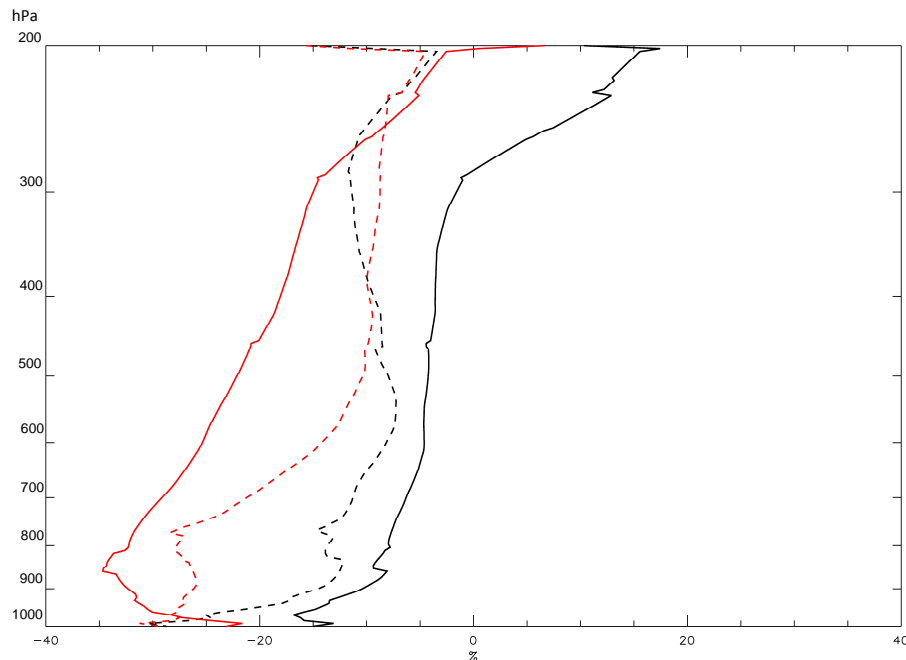


Fig. 2. Mean CO bias in % of modelled minus MOZAIC profiles. Black: MACC reanalysis. Red: control simulation without data assimilation. The solid lines show the means for NH extratropical airports (north of 30° N), the dashed lines the means for tropical airports (30° S–30° N). Data are averaged over the period January 2003 to December 2010.

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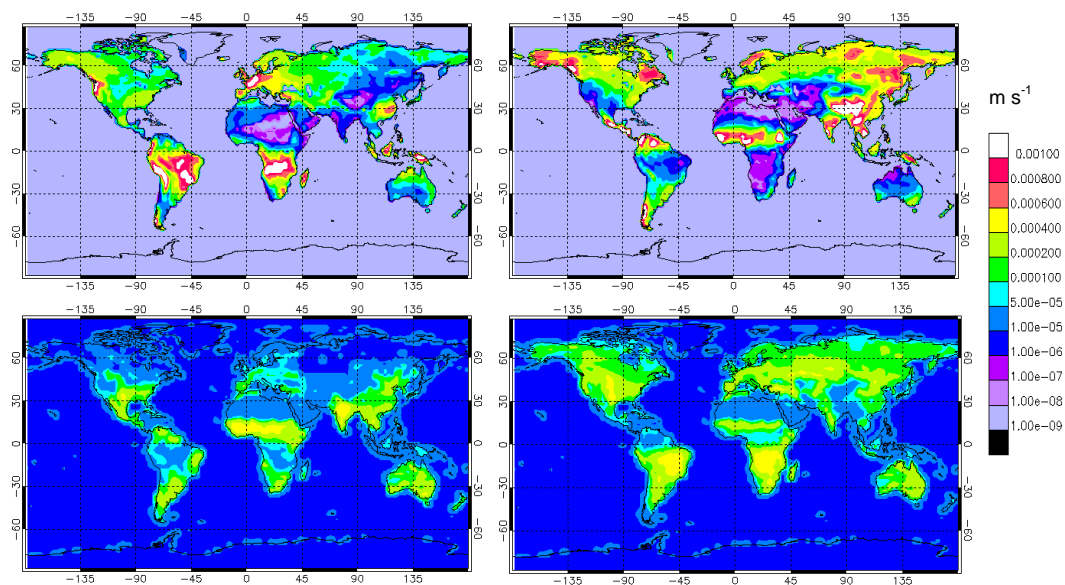


Fig. 3. Monthly mean dry deposition velocities in m s^{-1} for January (left) and July (right). Top panels: data derived with the ECHAM5/MOZ parameterization, bottom panels: data derived with the parameterization from Sanderson et al. (2003).

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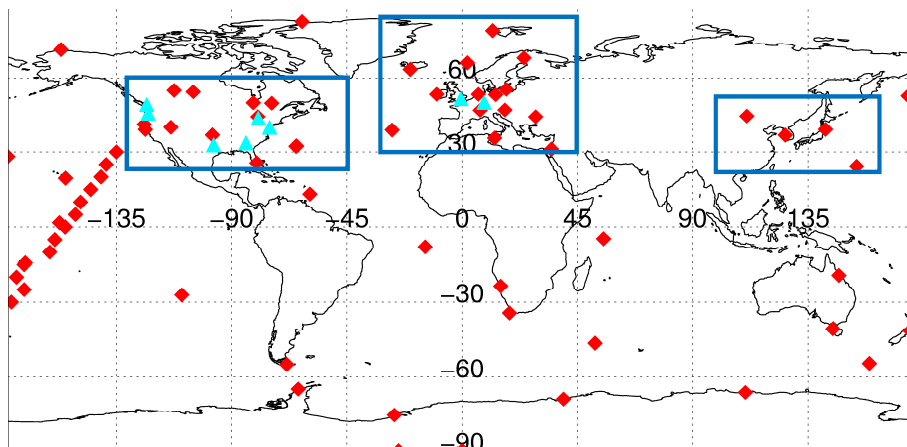


Fig. 4. Locations of the surface stations (red diamonds) and MOZAIC airports (turquoise triangles) used for the model evaluation. Selected evaluation regions are marked with blue boxes.

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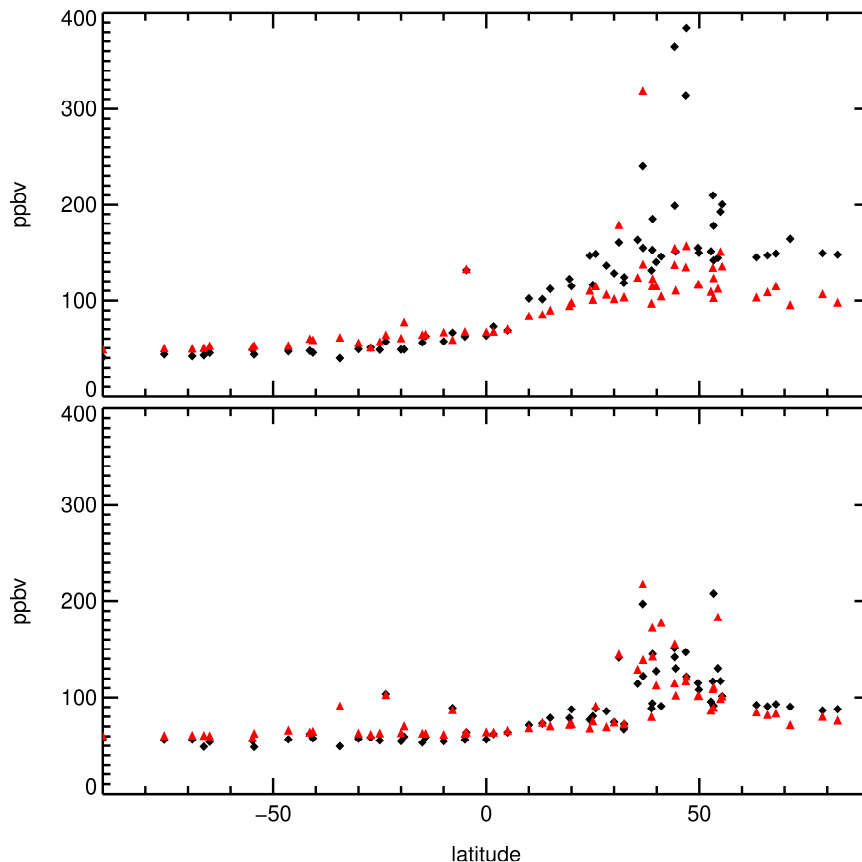


Fig. 5. Monthly mean CO mixing ratios from the 67 surface stations available in WDCGG used for this study sorted for geographical latitude (black rhombi) and modelled near-surface mixing ratios from simulation MI for the same locations (red triangles). Upper panel: January 2008; Lower panel: July 2008.

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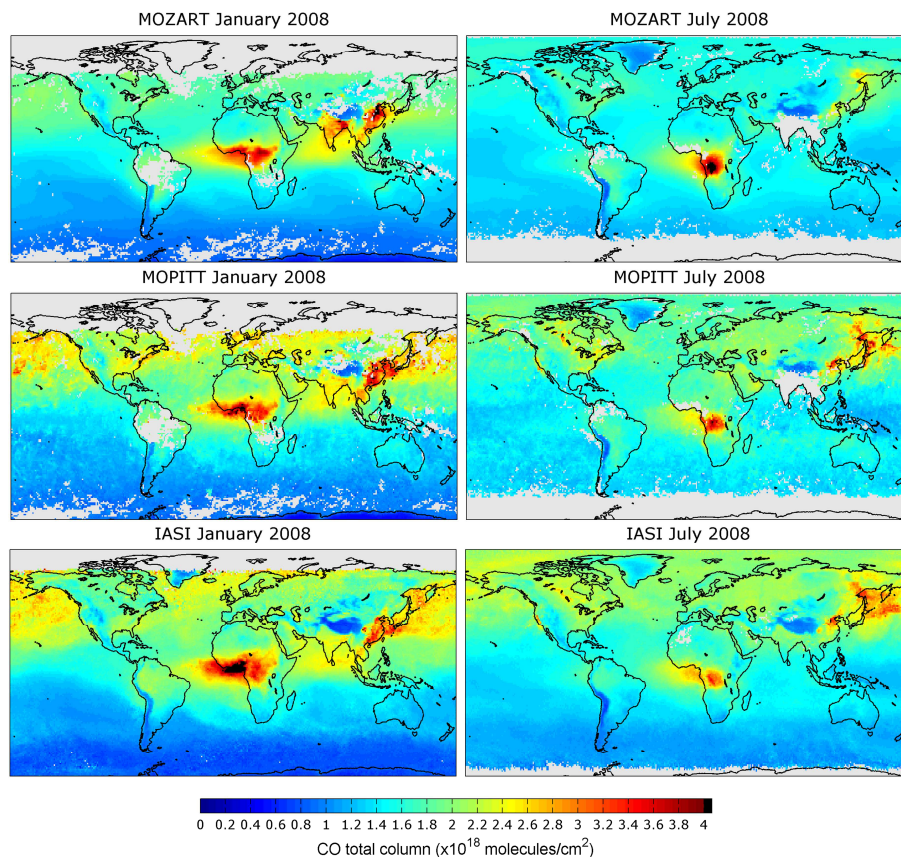


Fig. 6. Monthly mean CO total column densities for January and July 2008 as calculated from simulation MI smoothed by MOPITT averaging kernels, and observed from MOPITT and IASI.

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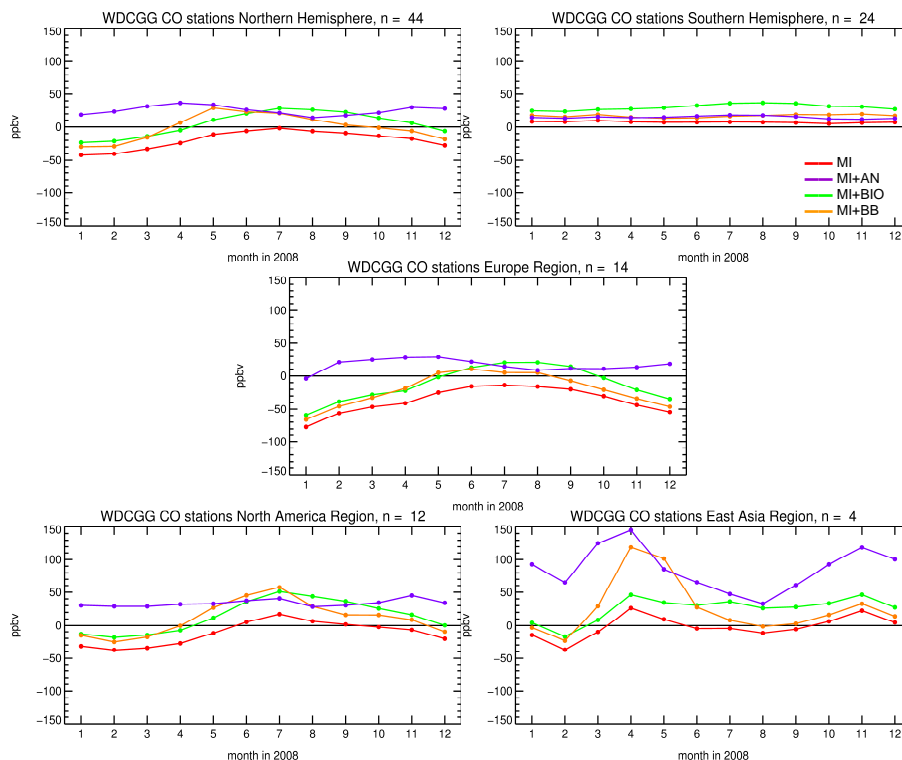


Fig. 7. Bias of modelled 2008 monthly mean surface level mixing ratios from the MOZART simulations MI, MI+AN, MI+BIO, and MI+BB compared to observations from WDCGG. n denotes the number of stations used.

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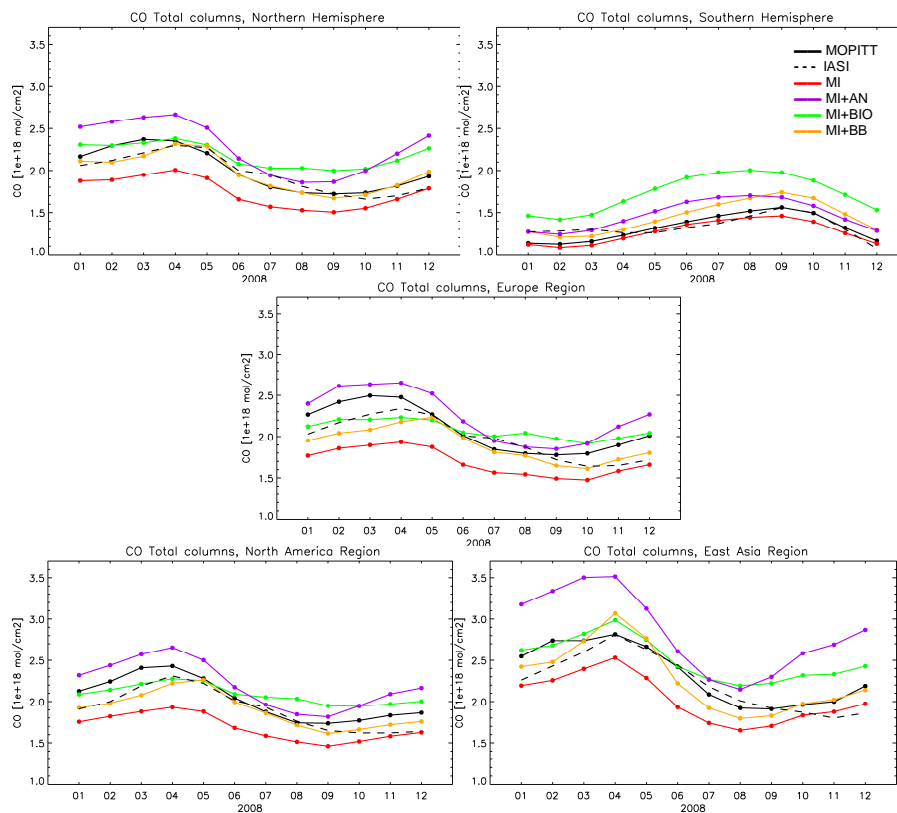


Fig. 8. CO total columns from MOPITT and IASI satellite observations compared to 2008 monthly modelled columns from the MOZART simulations MI, MI+AN, MI+BIO, and MI+BB.

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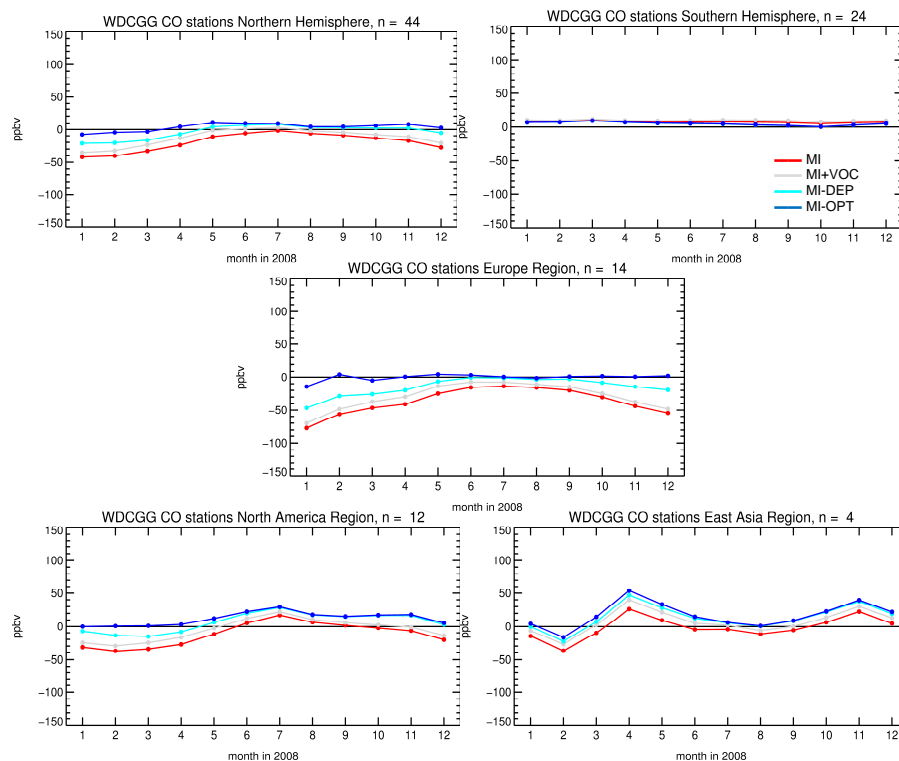


Fig. 9. As Fig. 7, but simulations MI (red), MI+VOC (grey), MI-DEP (light blue), and MI-OPT (dark blue).

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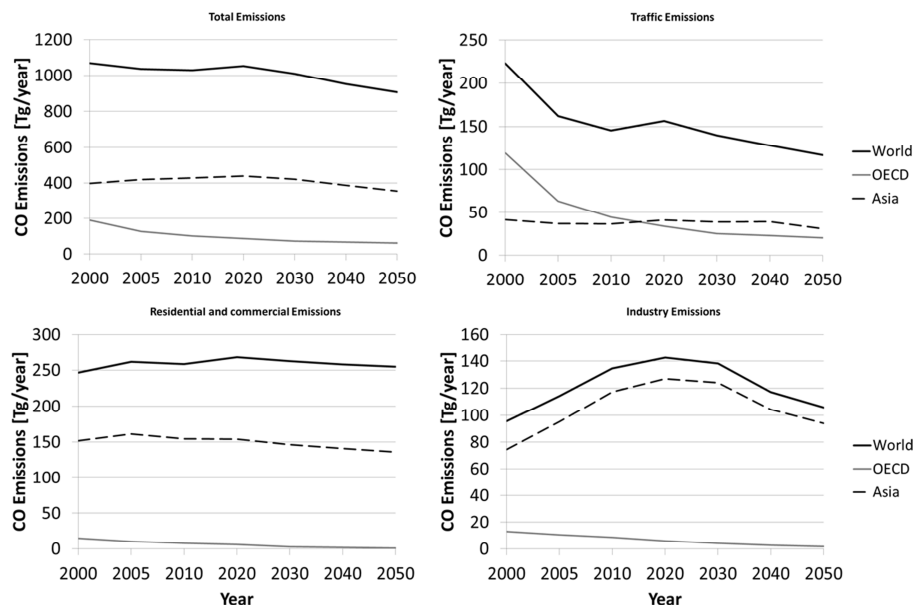


Fig. 10. Annual CO anthropogenic emissions 2000–2050 from different sectors in the RCP8.5 scenario, world-wide, for OECD countries and for Asia.

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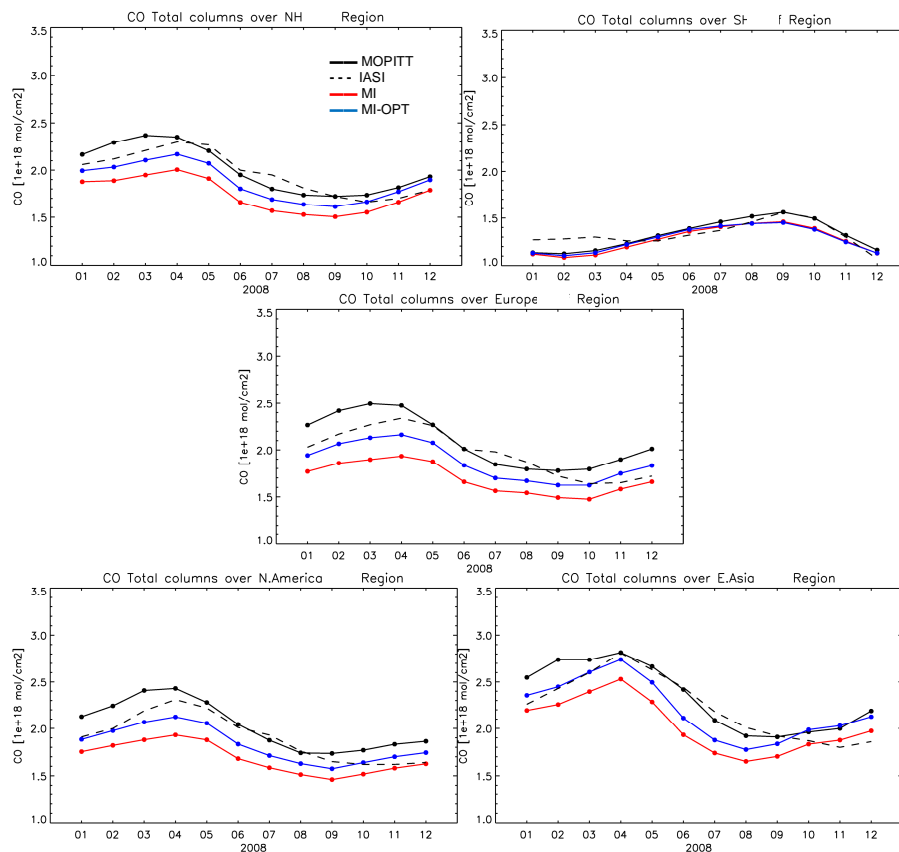


Fig. 11. As Fig. 8, but simulations MI (red) and MI-OPT (dark blue).

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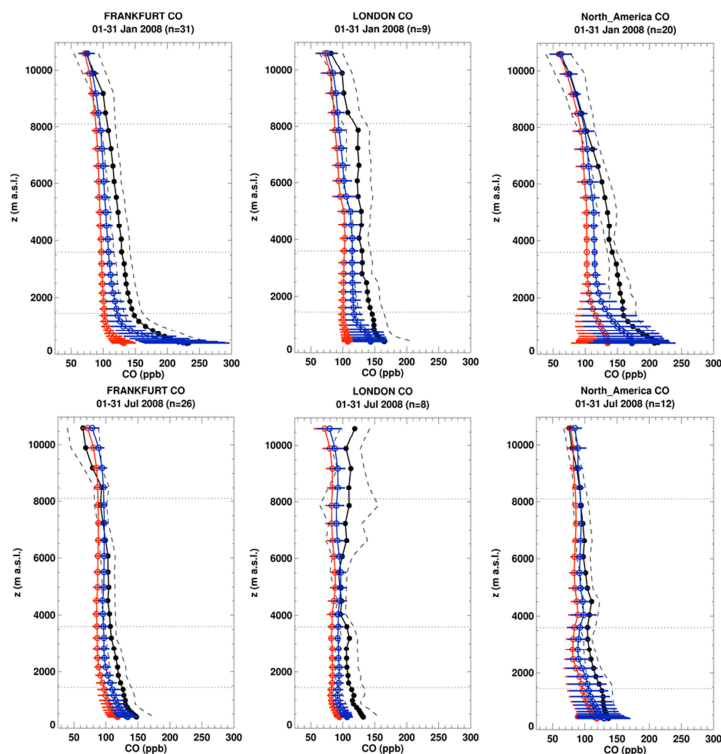


Fig. 12. Monthly averaged CO profiles from MOZAIC ascents and descents at selected airports evaluated at model levels (black solid lines). North_America composes of 6 airports in the United States and Canada, n is the total number of flights. The coloured lines depict model results interpolated to the same time and position from the simulations MI (red) and MI-OPT (blue). Horizontal lines give the range of the model results, black dotted lines the range of the MOZAIC observations.

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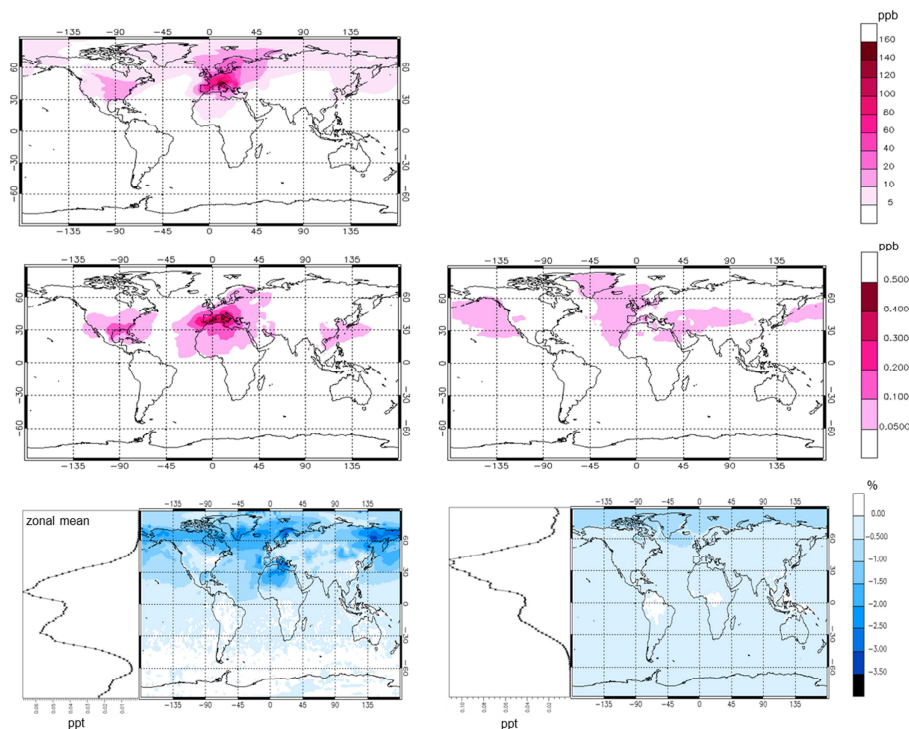


Fig. 13. Bias (MI-OPT – MI-DEP) of monthly mean surface level mixing ratios for January (left) and July (right) 2008. Top panels: Surface level CO absolute bias in ppb. Middle panels: Surface level ozone absolute bias in ppb. Bottom panels: OH relative bias in % and absolute MI-DEP zonal mean in ppt at 700 hPa. July CO differences are always lower than 5 ppb and are therefore not shown.